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(54) **Spherical reactor having a plurality of cylindrical reaction chambers and method for carrying out a reaction using said spherical reactor**

(57) Disclosed herein is a spherical or spheroidal reactor adapted to bring a feed gas into contact under elevated pressures with a fixed bed of a granular catalyst so that the feed gas is caused to undergo a chemical reaction to obtain a gaseous reaction product. The reactor has a spherical or spheroidal and pressure-resistant outer shell (3) and at least two cylindrical, intercylinder, truncated conical and/or truncated intercone reaction chambers (31, 32, 33), one inside the other, enclosed in the pressure-resistant outer shell. The reactor may carry a cylindrical protrusion.

FIG. 2

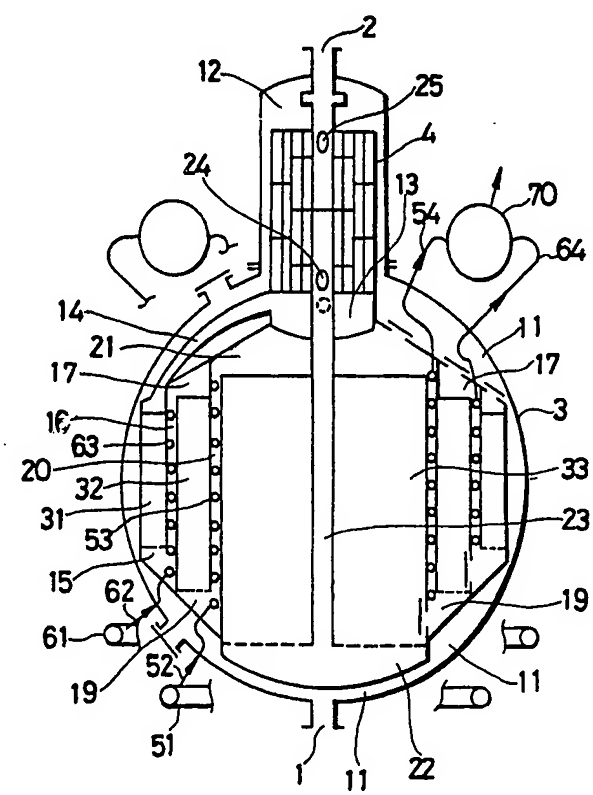

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FIG.1

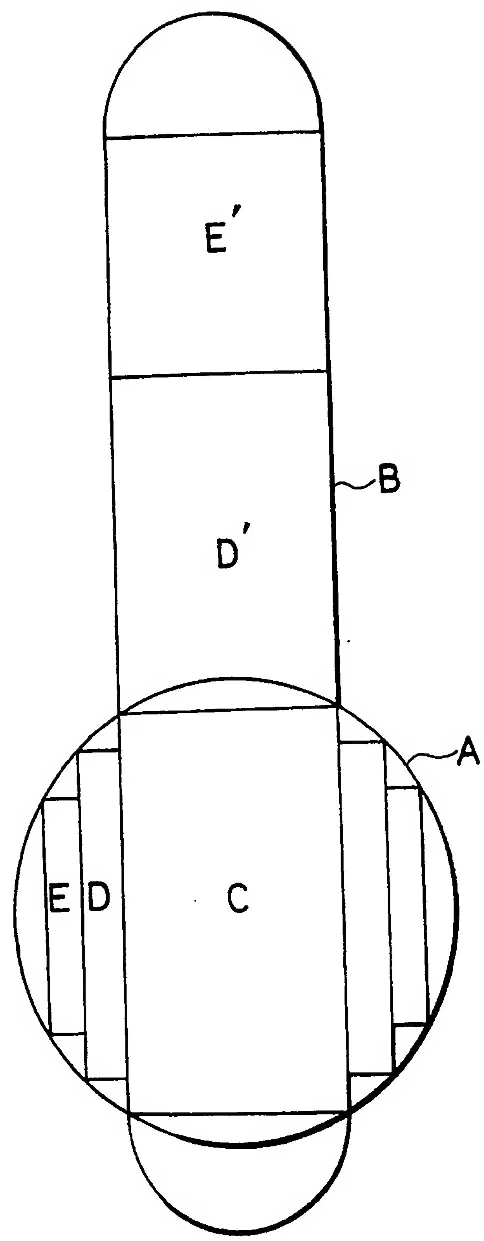

FIG. 2

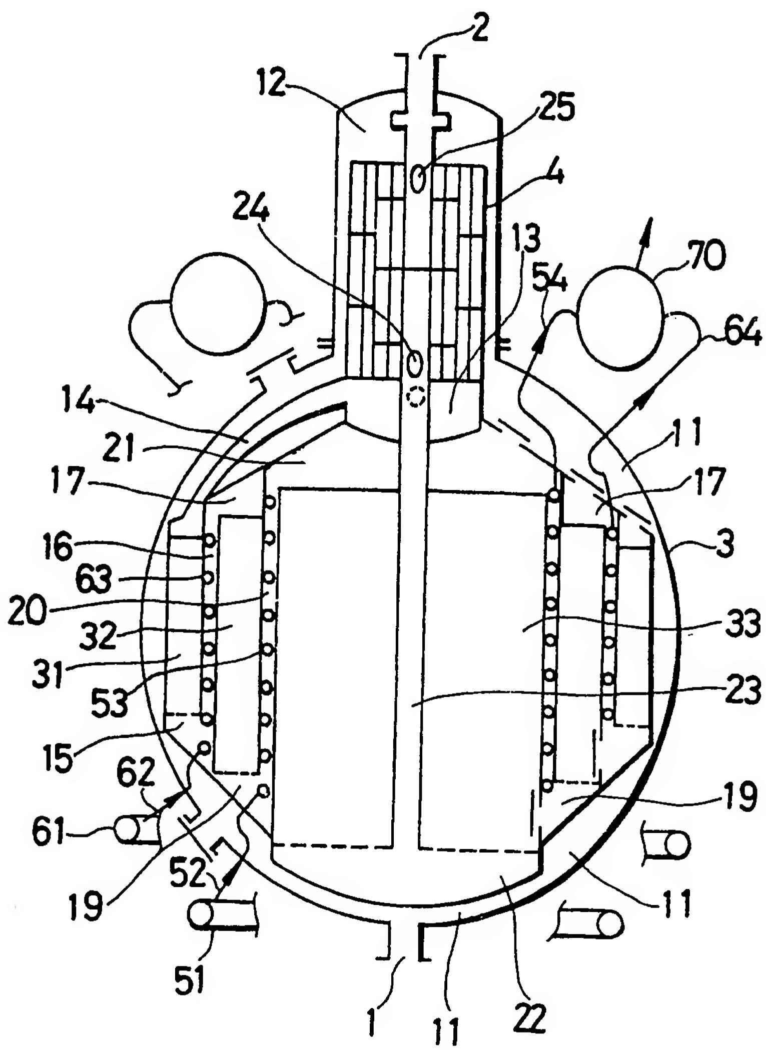
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FIG.3

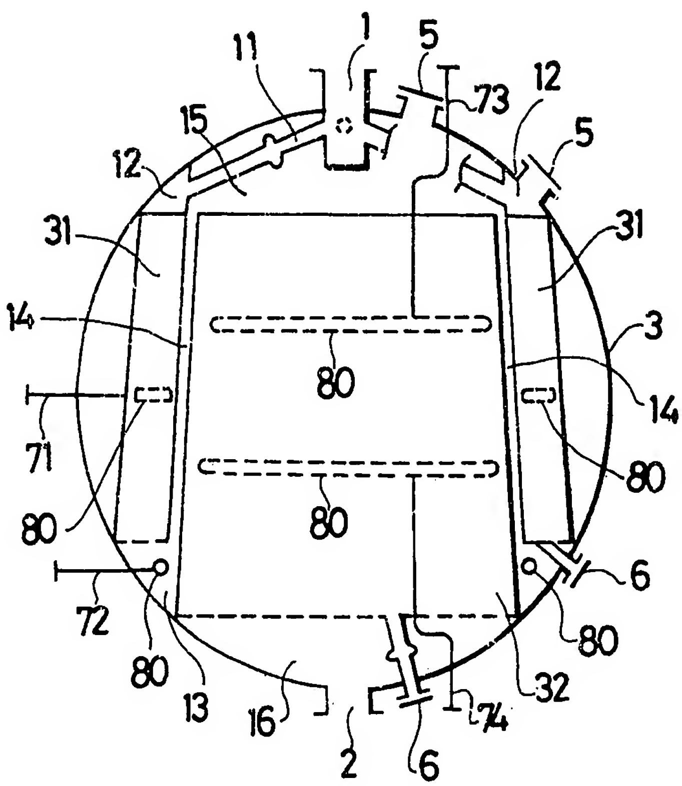


FIG.4

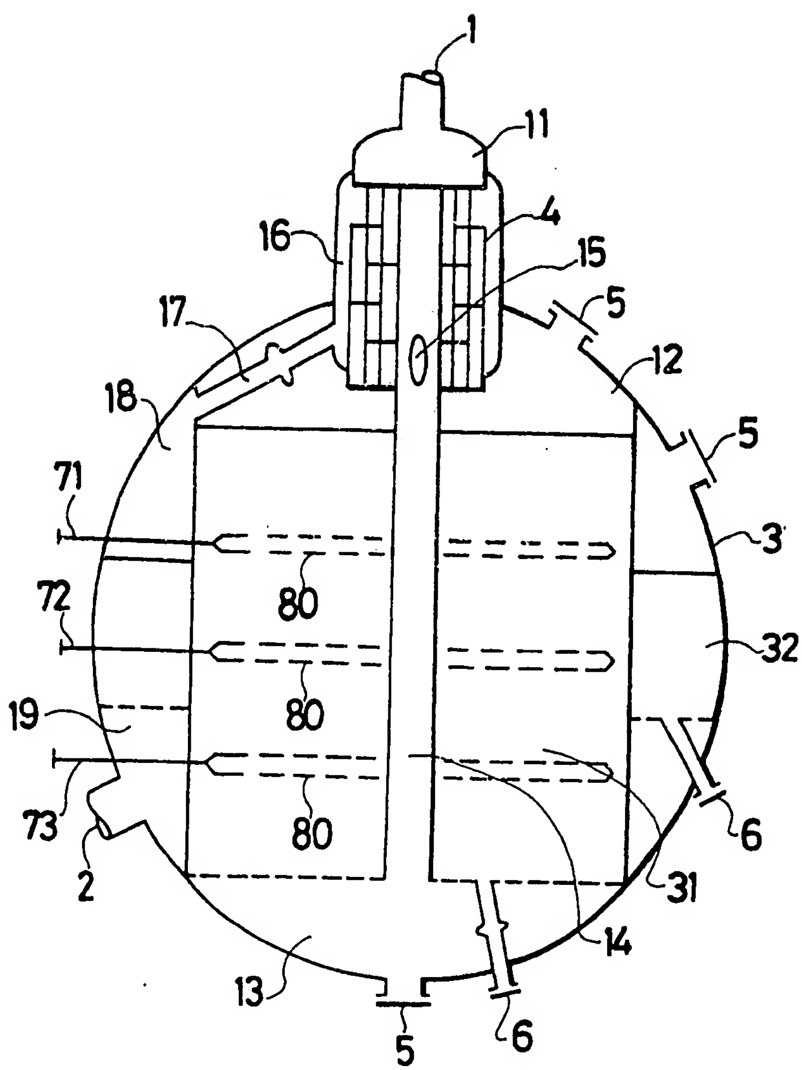
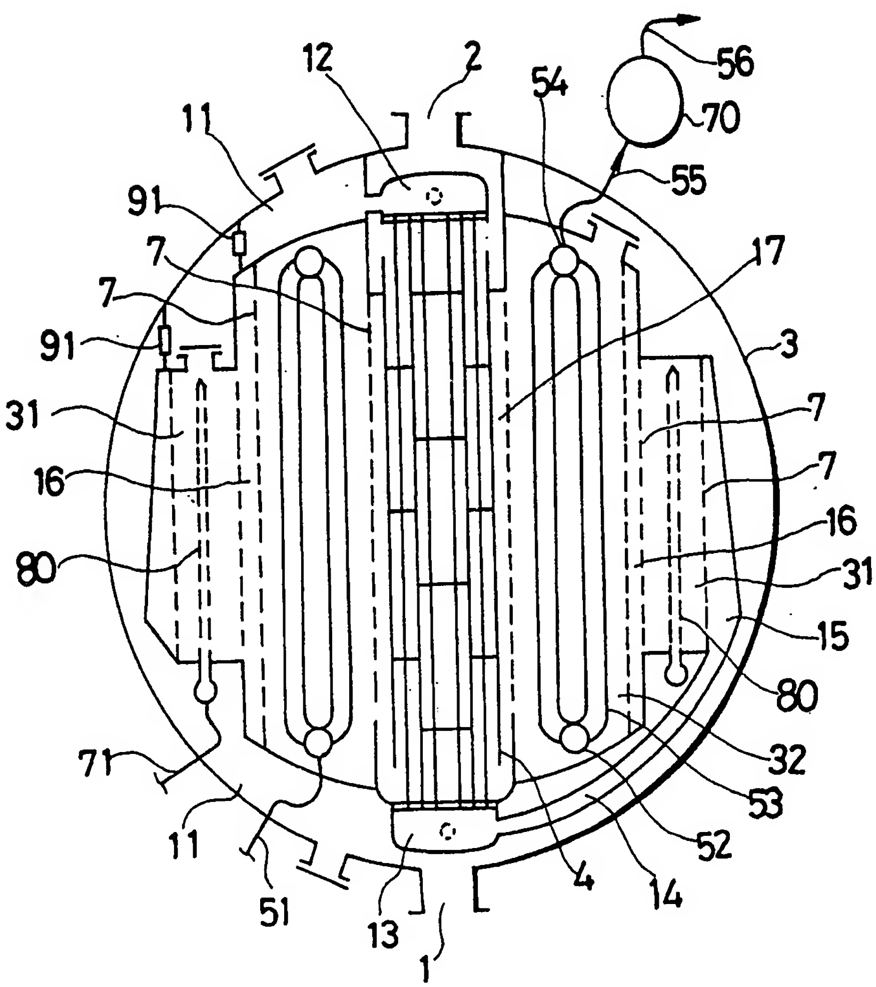


FIG. 5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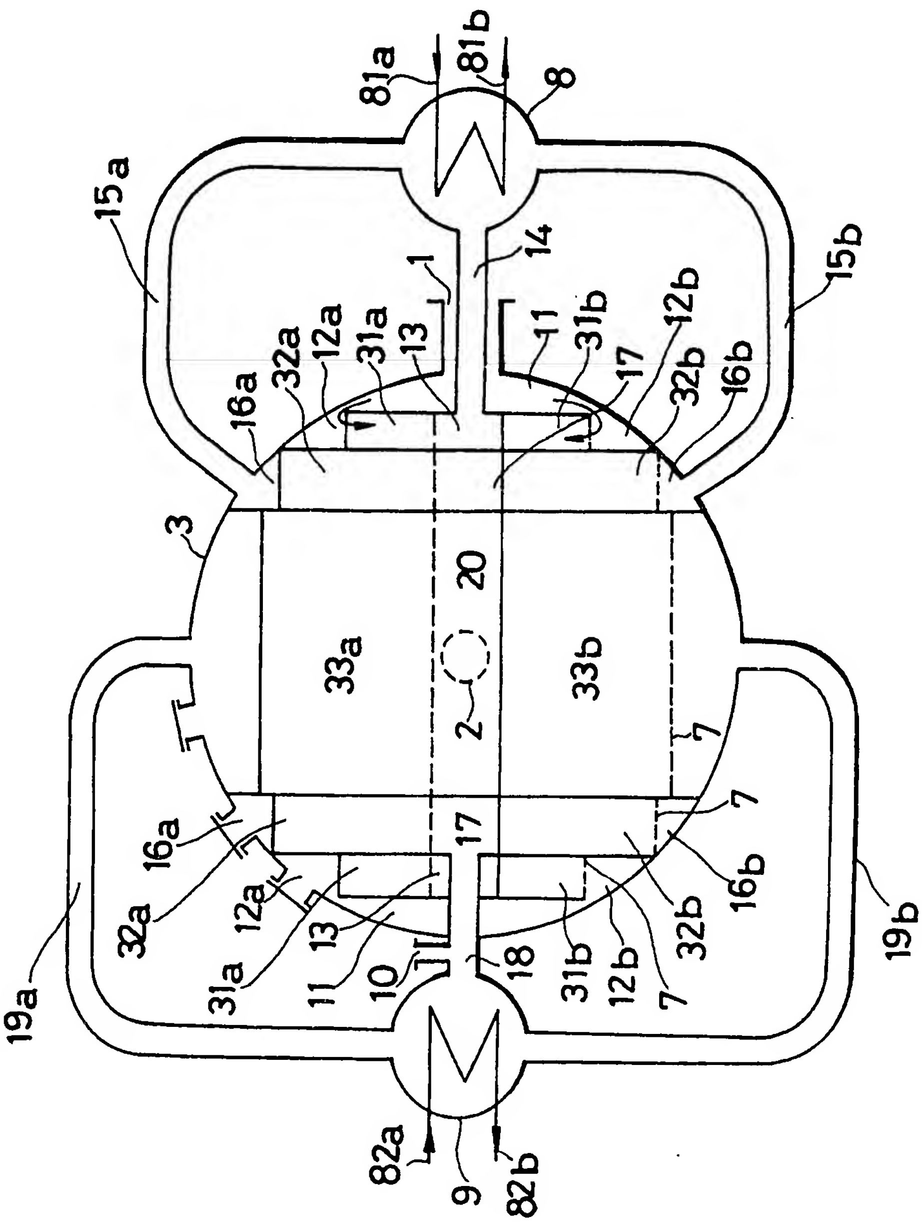

FIG. 6

SPECIFICATION

Spherical reactor having a plurality of cylindrical reaction chambers and method for carrying out a reactor using said spherical reactor

This invention relates to an improvement in or relating to a reactor adapted to cause a feed gas to undergo a chemical reaction under elevated pressures and using a fixed bed of a granular catalyst so that a gaseous reaction product can be obtained. More specifically, this invention relates to a reactor having a spherical or substantially spherical (hereinafter referred to as "spheroidal") and pressure-resistant outer shell. It also relates to a method for carrying out a reaction using such a spherical or spheroidal reactor.

As have already been put in operation in a number of ways led for example by the production of ammonia, methanol and the like, it has been routinely carried out to bring a high pressure gas into contact with a fixed bed of a granular catalyst (hereinafter merely called "catalyst") at the working temperature of the catalyst so that the gas is caused to undergo a chemical reaction, thereby obtaining a gaseous reaction product. A huge number of examples have already been disclosed as reactors for effecting such a chemical reaction. Many of such known reactors are each provided, in order to cool down its catalyst and gas which have been heated up as a result of exothermic reaction, with one or more openings for supplying a feed gas of a low temperature directly to one or more desired locations in a reaction chamber, which is present within the same reactor and packed with the catalyst and may optionally be divided into two or more sections, or to a location between divided reaction chambers; and/or one or more heat transfer surfaces adapted to effect an indirect heat exchange with the feed gas of the low temperature or a coolant other than the feed gas.

In such conventional reactors, each reaction chamber which is provided within its respective reactor and packed with a catalyst for its use is generally formed into a cylindrical or intercylinder shape, principally, from the viewpoints of establishing a uniform passage of a gas through the catalyst bed and facilitating its fabrication. Incidentally, the term "intercylinder shape" as used herein means a shape defined between two cylinders which have different diameters and are arranged coaxially, in other words, by the outer circumferential surface of the inner cylinder and the inner circumferential surface of the outer cylinder. As the flowing direction of a gas through each of these reaction chambers, it has been known to cause the gas to flow in the axial direction of the cylindrical or intercylinder reaction chamber (i.e., axial flow) or in the radial direction of the same reaction chamber (i.e., radial flow). Accordingly, a cylindrical outer shell provided with lids at both ends thereof has been used, in a conventional reactor, as a pressure-resistant outer shell for enclosing a cylindrical or intercylinder reaction chamber therein. To the best knowledge of the present inventors, no pressure-resistant outer shell has ever been proposed other than such cylindrical pressure-resistant outer shells.

The principal object of this invention is to reduce the wall thickness of a pressure-resistant outer shell of a reactor so as to lower the weight of the reactor; and to make the reactor shorter so as to shorten the lengths of accessory pipings, thereby making a support and foundation required for the installation of the reactor smaller and thus cutting down the construction cost of the reactor.

As mentioned above, the present invention relates to a reactor having a spherical or spheroidal and pressure-resistant outer shell. In case the internal volume and internal gas pressure are the same, a spherical pressure-resistant vessel has a smaller surface area and permits use of thinner steel material for its pressure-resistant outer shell, compared with a cylindrical pressure-resistant vessel. For example, in case a spherical pressure-resistant vessel and cylindrical pressure-resistant vessel have the same inner diameter and are used at the same pressure, the thickness of a steel material for the former vessel may be reduced to about one half of that of a steel material required for the latter vessel. Correspondingly, it has been well known that the weight of a pressure-resistant outer shell *per se* may be rendered lighter when its shape is changed to a spherical one. Making use of this advantage, many spherical pressure-resistant vessels are used, for example, as storage tanks for liquefied petroleum gas. However, nothing has been reported regarding how the internal space is utilized when a spherical pressure-resistant vessel is used as the outer shell of a pressure-resistant reactor.

The present inventors have carried out an investigation on the utilization of the internal space of a spherical pressure-resistant outer shell as a reactor. As a result, the present invention has been completed.

In one aspect of this invention, there is thus provided a reactor adapted to bring a feed gas into contact with a fixed bed of a granular catalyst under elevated pressures so that the feed gas is caused to undergo a chemical reaction and a gaseous reaction product is thus obtained, said reactor comprising a pressure-resistant outer shell and at least two reaction chambers enclosed within the outer shell; the outer shell is either (a) a sphere or a spheroid formed of a cylinder having a length equal to or shorter than the three quarters of its diameter and hemispherical end plates having the same diameter as the cylinder and

provided at both ends of the cylinder or (b) another sphere or spheroid having the same configurations as the former sphere or spheroid and provided with at least one cylindrical protrusion which covers a quarter of the overall surface area of the latter sphere or spheroid or less; and the reaction chambers are of cylindrical, intercylinder, truncated conical and /or truncated intercone shapes and/or shapes defined respectively between parts or the entire parts of the outer surfaces of the cylindrical, intercylinder, truncated conical or truncated intercone shapes and the inner surface of the outer shell and having annular cross-sections when taken along planes perpendicular to the axes thereof and are arranged in such relationship that the reaction chamber having a smaller outer diameter is present inside the reaction chamber having a greater inner diameter.

In another aspect of this invention, there is also provided a method for bringing a feed gas into contact with a fixed bed of a granular catalyst under elevated pressures so that the feed gas is caused to undergo a chemical reaction and a gaseous reaction product is thus obtained, which method is characterized in that the reaction is proceeded by using the above-described reactor and causing the feed gas to pass successively through at least two reaction chambers in the reactor.

Embodiments of the present invention will now be described, by way of example, with reference to the accompanying drawings, in which:—

Figure 1 is a schematic cross-sectional view of each of a cylindrical reactor and spherical reactor, illustrating the principle of this invention;

Figure 2 is a schematic cross-sectional view of one embodiment of the reactor according to this invention, in which the spherical outer shell is provided with a cylindrical protrusion and the spherical shell encloses one cylindrical reaction chamber and two intercylinder reaction chambers;

Figure 3 is a schematic cross-sectional view of another embodiment of the reactor according to this invention, in which the outer shell is formed of a sphere and contains two reaction chambers, one resembling a frustum of a cone and the other having a truncated intercone shape;

Figure 4 is a schematic cross-sectional view of a further embodiment of the reactor according to this invention, in which the outer shell is a combination of a sphere and a cylindrical protrusion and encloses two reaction chambers, one being cylindrical and the other substantially toroidal;

Figure 5 is a schematic cross-sectional illustration of a still further embodiment of the reactor according to this invention, in which the outer shell is made of a sphere and encloses two intercylinder reaction chambers and a feed gas is caused to flow radially through the reaction chambers; and

Figure 6 is a schematic cross-sectional view of a still further embodiment of the reactor according to this invention, in which the outer shell is formed of a sphere and contains one cylindrical reaction chamber and two intercylinder reaction chambers, and the reactor is provided with external heat exchangers.

The fundamental characteristic of using the internal space of a spherical and pressure-resistant outer shell as a reactor in accordance with this invention resides in the provision of at least two reaction chambers in the internal space, which reaction chambers are individually of cylindrical, intercylinder, truncated conical and/or truncated intercone shapes and/or toroidal shapes defined respectively between parts or the entire parts of the outer surfaces of the cylindrical, intercylinder, truncated conical or truncated intercone shapes and the inner surface of the outer shell. Owing to the above fundamental characteristic, the internal space of a spherical and pressure-resistant outer shell can be effectively utilized for a reactor and the weight of the reactor can be reduced compared with any prior art cylindrical reactor. The basic features of this invention will first be explained with reference to Fig. 1.

Fig. 1 is a schematic cross-sectional view for comparing the surface areas of a spherical outer shell *A* with that of a cylindrical outer shell *B* which encloses three reaction chambers having the same horizontal cross-sectional area. Letter *C* indicates a cylindrical reaction chamber placed in contact with the inner surface of the spherical reactor *A* and adapted to hold a catalyst in a packed state therein. According to the illustration, the reaction chamber *C* is common to both of the outer shells. Letter *D* is an intercylinder reaction chamber which is placed in contact with the inner surface of the spherical reactor *A* outside the reaction chamber *C* and has the same horizontal cross-sectional area as the reaction chamber *C*. Designated at letter *D'* is another cylindrical reaction chamber which has the same internal volume and horizontal cross-sectional area as the reaction chamber *D* and is mounted on the reaction chamber *C*. The internal volume of the reaction chambers *D* and *D'* is smaller than that of the reaction chamber *C*. Letter *E* is a further intercylinder reaction chamber which is placed in contact with the inner surface of the spherical reactor *A* outside the reaction chamber *D* and has the same horizontal cross-sectional area as the reaction chambers *C* and *D*. Letter *E'* is a further cylindrical reaction chamber which is equal in internal volume and horizontal cross-sectional area to the reaction chamber *E* and is mounted on the reaction chamber *D'* within the cylindrical reactor *B*. The volume of each of the reaction chambers *E* and *E'* is smaller

than that of each of the reaction chambers *D* and *D'*. Owing to the above arrangement, the spherical reactor *A* can be packed with a catalyst of the same volume as the cylindrical reactor *B*. Furthermore, the linear velocity to be achieved by a gas passing through each of the reaction chambers *C, D* and *E* will become equal to the linear velocity of a gas passing axially through the cylindrical reactor *B* when a feed gas is caused to flow axially in the order of C→D→E or E→D→C through the spherical reactor *A*. Accordingly, the spherical reactor *A* is equal to the cylindrical reactor *B* in both catalyst volume and pressure loss to be developed upon passage of a gas therethrough. When one attaches known lids to both upper and lower ends of the cylindrical reactor *B* (hemispherical end plates in the embodiment illustrated in Fig. 1) and calculates the surface areas of both reactors by a method known *per se* in the art, it is readily understood that the surface area of the spherical reactor *A* is considerably smaller than that of the cylindrical reactor *B*. Assuming that both reactors are respectively charged with high pressure gases of the same pressure, the pressure-resistant outer shell of the spherical reactor can be made 10–20% lighter than that of the cylindrical reactor or even lighter even if the additional weight of a below-described cooling system to be provided between the reaction chambers is taken into consideration. Such a weight decrease of a pressure-resistant outer shell is achieved by making its wall thickness thinner. The upper limit of a large reactor is frequently restricted by the maximum thickness of its steel material which is produceable. The decrease in required wall thickness means that such a limitation can be successfully avoided.

The above description pertains to the principle of this invention. Besides cylindrical or intercylinder reaction chambers, it is possible to use, in the present invention, reaction chambers of truncated conical or truncated intercone shapes which have never been employed in any conventional cylindrical reactors. Truncated conical and truncated intercone reaction chambers will be described later in the patent specification. By the way, the reactor according to this invention may be used for the practice of two types of chemical reactions, i.e., both exothermic and endothermic reactions. Reference to an endothermic reaction will be made later and the present invention will hereinafter be described with principal reference to exothermic reactions. It should however be noted that there is no basic difference between these two types of reactions so long as this invention is concerned and the following description can be applied equally to an endothermic reaction by reversing the thermal aspects other than the preheating of each feed gas, for example, by reading the cooling in an exothermic reaction as the heating in an endothermic reaction. It is generally necessary to provide, within the interior of a reactor, devices and equipments adapted to cool its catalyst or gases, which have been heated as a result of the reaction, to a desired temperature and to preheat its feed gas to the working temperature of the catalyst, in addition to the above-described reaction chambers. As the first method of cooling down the temperatures of the catalyst and hot gas in the interior of a reactor, it is possible to supply the feed gas of a low temperature directly to one or more desired locations within the catalyst bed or, where the catalyst bed (i.e., the reaction chamber) is divided, to the gas flow passage connecting an upstream-side catalyst bed and its subsequent downstream-side catalyst bed. As the second method for achieving the same objective, it is feasible to provide a heat transfer surface for indirect heat exchange in the catalyst bed or, in case the catalyst bed is divided, in a space between an upstream-side reaction chamber and its subsequent downstream-side reaction chamber so as to subject hot catalyst or gas to heat exchange with the feed gas of the low temperature. The second method may also be effective to preheat the feed gas. In addition, as the third method for the same objective, it is also possible to use, in lieu of the feed gas which serves as the low-temperature side fluid in the heat exchanger in the second method, another cooling fluid, for example, another gaseous medium or a liquid or mixed-phase fluid consisting of a liquid and its vapour, which liquid or mixed-phase fluid has reached its boiling point at a given temperature under an elevated pressure. The feed gas may be preheated in accordance with the above-mentioned second method or by subjecting it to indirect heat exchange with a hot gas flowing out of the most downstream reaction chamber prior to introducing the feed gas into the reactor. In case a pressure-resistant outer shell does not encounter any problems or inconvenience in maintaining its strength and avoiding its corrosion and embrittlement even if the pressure-resistant outer shell is heated up to the working temperature of its catalyst, it is possible, as a method for preheating the feed gas, to heat the feed gas in a heat exchanger provided outside the reactor and then to introduce the feed gas, which has been heated to the working temperature of the catalyst, into the reactor. It is however necessary to provide the above-described indirect heat exchanger, which is adapted to preheat each feed gas, within a reactor if it is required to hold the temperature of the pressure-resistant outer shell below the working temperature of the catalyst.

According to this invention, the interior of a spherical and pressure-resistant outer shell is utilized as a space for enclosing reaction chambers as well as functional equipments for

the cooling of a catalyst and gas and the preheating of a feed gas as described above. The volume required for the installation of each of these equipments varies to a considerable extent depending on the kind of a reaction which takes place in the reaction chambers, the type of a catalyst used, the heat quantity generated upon occurrence of the reaction, reaction temperature, reaction pressure and other reaction parameters. Therefore, the present invention can bring out, besides the above-described weight and wall thickness reduction of each reactor, many other merits. Among such merits, those common to many of embodiments of this invention will first be described.

The advantageous features of this invention can not be clearly exhibited generally in reactions employing a reaction pressure of 5 kg/cm^2 or lower (in terms of gauge pressure; all designations of kg/cm^2, which will follow, refer to kg/cm^{2}G unless specified otherwise), because there is no substantial difference between the weight of a steel material required for the pressure-resistant outer shell of a spherical reactor according to this invention and that of a cylindrical reactor using the same amount of a catalyst as the spherical reactor. The advantages of this invention become clearer at reaction pressures of 10 kg/cm^2 and higher, and particularly, at reaction pressures of 30 kg/cm^2 and higher. With respect to reaction chambers, substantially similar to cylindrical reactors, it is preferable to use cylindrical and/or intercylinder reaction chambers for the spherical reactor according to this invention so that a gas is allowed to pass through each reaction chamber uniformly in a cross-section perpendicular to the flowing direction of the gas and the effective utilization rate of the catalyst can be increased. However, in case only a single cylindrical or intercylinder reaction chamber is provided in a spherical reactor, there remains a large internal space other than the reaction chamber. Even if the equipment for cooling the catalyst and gas and the equipment for preheating the feed gas are enclosed in the internal space, the spherical reactor still has a surplus space therein and the advantages of the spherical reactor are not brought about. Accordingly, it is indispensible to provide at least two cylindrical or intercylinder reaction chambers in order to make an effective use of the internal space of each spherical reactor. Different from cylinder reactors, it is readily possible, for the effective utilization of the internal space of each spherical reactor, to employ, besides cylindrical and intercylinder reaction chambers, truncated conical reaction chambers, truncated intercone reaction chambers, as well as to use as reaction chambers substantially toroidal spaces formed between parts or the entire parts of the outer surfaces of such cylindrical, intercylinder, truncated conical and truncated intercone shapes and a part of the inner surface of its pressure-resistant outer shell. Use of reaction chambers of such various configurations is in fact advantageous. This is apparent from the fact that a spherical reactor is short and thick. Therefore, the term "at least two reaction chambers" as used herein embraces reaction chambers other than cylindrical or intercylinder reaction chambers as described above. Merits of these reaction chambers other than cylindrical or intercylinder reaction chambers will be described with reference to their specific embodiments. It is also important for the effective utilization of the internal space of each spherical reactor to arrange these two or more reaction chambers of the above-described shapes in such a way that they share the common axis, in other words, they are coaxial and the reaction chamber having a smaller outer diameter is placed inside the reaction chamber having a greater inner diameter.

The first structural merit of a spherical reactor having such a structure as described above is that the wall thickness and weight of its outer shell can be reduced compared with a cylindrical reactor having the same catalyst packing volume. The reasons for the first structural merit have already been described above. The second structural merit of the spherical reactor is that connector openings, which are adapted to charge or discharge a gas, a cooling medium or catalyst, can be formed with ease at desired locations in the spherical surface of the spherical and pressure-resistant outer shell. It is of course possible to provide a space for charging or discharging a gas or cooling medium at a location remote from both end plates of a conventional reactor having an elongated cylindrical shape. However, this requires a pressure-resistant outer shell having a considerably elongated length. Particularly in case an inner cylinder is required as a reaction chamber, a substantial thermal expansion or contraction difference occurs between the outer shell and the inner cylinder due to temperature difference developed during each operation. Therefore, it makes the overall structure complex and is thus impractical to provide tubes through both circumferential wall of the cylindrical and pressure-resistant outer shell and circumferential wall of the inner cylinder for charging or discharging gas and cooling medium. Contrary to the conventional cylindrical reactor, the spherical reactor according to this invention makes use of the toroidal space formed within the reactor by a division wall and the inner spherical surface of the pressure-resistant outer shell and permits to provide tubes for charging or discharging a gas, cooling medium and the like as bent tubes or coils extending along the inner surface of the pressure-resistant outer shell. This permits to make the structure simpler and avoids devel-

opment of large thermal stresses due to the thermal expansion of the reactor. Furthermore, the provision of the above-described toroidal space is convenient or advantageous for the fabrication of a large reactor, because it is wide enough to permit workers to work therein.

In the above description of the principle of the present invention which description was made with reference to Fig. 1, a circumferential wall of a reaction chamber is disposed in contact with its adjacent circumferential wall of another reaction chamber in order to make the description simpler. It is certainly desirous to dispose reaction chambers in such a way as their circumferential walls are partly or mostly kept in mutual contact as shown in Fig. 1, since such an arrangement of reaction chambers permits to use more space in the spherical reactor as reaction chambers. However, it becomes necessary to provide a space for the installation of such a cooling system as described above within each reactor irrespective of the shape of the reactor, in case a large heat quantity is produced in the course of a reaction which takes place in its reaction chamber, the catalyst packed within the reaction chamber has low resistance to heat and its capacity is lowered by any excessive temperature increase, or raised gas and catalyst temperatures adversely affect on the progress of the reaction in view of its chemical equalibrium. When forming a space in a conventional cylindrical reactor to provide the above-described cooling system, namely, a feed line of a low temperature feed gas to a reaction chamber or to a gas flow passage between two adjacent reaction chambers or a tubular heat transfer surface for subjecting a high temperature gas to indirect heat exchange with the low temperature feed gas, without changing the amount of its catalyst, it becomes necessary to increase the diameter or length of the cylindrical reactor. On the other hand, in a spherical reactor having two or more reaction chambers, a toroidal space is left between the reaction chamber and the outer shell of the spherical reactor. The thus-left space can be used directly as a flow passage for a reaction gas or as an installation space for tubes adapted to cause the reaction gas, feed gas and cooling medium to pass therethrough, thereby increasing the utilization rate of the internal space without increasing the internal volume of the spherical reactor.

As apparent from the above description, it is unnecessary for the spherical reactor according to this invention to have an exactly spherical configuration in order to bring about the merits of this invention. The merits of this invention can still be obtained by reactors which have configurations similar to a sphere. Even if a part of a spherical outer shell is protruded outwardly, the merits of this invention are not adversely affected so long as the protrusion is cylindrical and the diameter of the cylindrical protrusion is relatively small compared with the diameter of the spherical outer shell. For instance, according to a result obtained by the present inventors with respect to a spherical outer shell provided with one or more cylindrical protrusions, the internal spaces of which protrusions are in communication with the internal space of the spherical outer shell, it has been found that substantially the same merits as a spherical reactor having no protrusion may be brought about even by a spherical reactor in which the surface area of the part—presenting a spherical surface with the joining part between the cylindrical protrusion and the spherical outer shell as its boundary—accounts for at least three quarters of the entire surface area of the same spherical outer shell without the cylindrical protrusion. In this case, the above-described merits can be substantially brought about even by a spherical reactor having a considerably long cylindrical protrusion, compared with a cylindrical reactor having the same internal space as the spherical outer shell of the spherical reactor. It is not necessary to limit the number of these cylindrical protrusions to only one but it is feasible to provide many small protrusions at desired locations on the surface of a spherical outer shell. As another example of reactors which example can also bring about the merits of this invention but does not present exactly spherical surface, may be mentioned a spheroidal reactor formed of a cylinder having a length not longer that the three quarters of its diameter and hemispherical end plates having the same diameter as the cylinder and provided at both ends of the cylinder. This spheroidal reactor is of a preferred shape as will be described later, when a large heat quantity is produced by the reaction, the gas is caused to flow at a high velocity through the reaction chamber and the provision of a wide heat transfer area is required for indirect cooling in the reaction chamber. In the above-described spheroidal reactor, the provision of one or more cylindrical protrusions does not hamper the achievement of the merits of this invention, similar to the spherical reactor. In the following description, the above-described spherical and spheroidal reactors will be called merely "spherical reactors" irrespective of the presence and the absence of cylindrical protrusions. Here, although it is preferable in many instances to arrange the common axis of each reaction chamber of a reactor in a vertical direction as shown in Fig. 1 for the assembly and the maintenance and inspection of the reactor, no functional problem or inconvenience will arise even if the common axis extends horizontally or aslant because the raw materials and reaction product are both gaseous.

Next, the operation of the spherical reactor according to this invention will be described, it is possible to provide, within the interior of the reactor of this invention, two or more reaction chambers having the same cross-sectional shape when seen along a plane perpendicular to the flowing direction of a gas and the same length along the flowing direction of the gas. When such two or more reaction chambers are packed with the same catalyst, a feed gas can be passed parallely through the two or more reaction chambers. However, such a parallel passage of a feed gas has a potential danger that the flow rate of the feed gas through one reaction chamber may not be equalized to that of the feed gas passing through another reaction chamber and the channelling phenomenon thus occurs. The danger of the channelling phenomenon becomes greater when the feed gas is caused to flow parallelly through reaction chambers having different cross-sectional shapes or different lengths. In order to avoid the channelling phenomenon, it is necessary to add certain means to each reaction chamber so as to maintain the same gas flow rate through the reaction chamber. On the other hand, the internal space of a spherical reactor may not be most effectively utilized by providing two or more reaction chambers having the same cross-sectional shape when seen along a plane perpendicular to the flowing direction of a gas and the same length in the flowing direction of the gas within the spherical reactor. For these reasons, it is most desirous to use the reactor according to this invention by causing a feed gas to pass successively through at least two reaction chambers provided within the spherical reactor. By causing the feed gas to pass successively through the reaction chambers as mentioned above, it becomes possible to pack a large amount of a catalyst within the spherical reactor, to effectively utilize the catalyst, and to increase the effective utilization rate of the internal space of the spherical reactor. The first merit, which can be obtained by causing a feed gas to flow successively through two or more reaction chambers, resides in that it becomes easier to avoid the overheating of a catalyst packed at the upstream side relative to the flow of the feed gas (particularly at the most upstream side). Namely, it is necessary to bring a feed gas, which has been preheated to the working temperature of a catalyst, into contact first of all with the catalyst packed at the most upstream side in order to make effective use of the catalyst. At the most upstream side, the feed gas does not contain the reaction product at all or contains only little reaction product. Accordingly, a violent reaction takes place in the catalyst bed packed at the most upstream side, resulting in abrupt increases of both gas and catalyst temperatures. It has been well known that such an excessive increase of the catalyst temperatures gives such deleterious effects as lowered catalytic activity and more occurrence of undesired by-products to many catalysts. These problems may be easily solved by using at the upstream side a reaction chamber having a smaller cross-sectional area when seen along a plane perpendicular to the flowing direction of the gas and a shorter length so as to maintain a high gas flow velocity in the reaction chamber and to control the reaction velocity and the development of too much reaction heat; and by cooling a gas leaving from the reaction chamber by any of the above-described cooling means. If the gas is then caused to pass at a lower velocity through one or more subsequent reaction chambers having greater cross-sectional areas as the concentration of the reaction product in the gas becomes higher, the reaction product can be obtained in exactly the same manner as in a conventional cylindrical reactor. At the same time, it is possible to obtain such an additional effect that the service life of the catalyst can be prolonged or the occurrence of by-products can be reduced. If one wants to obtain the above-described effects in a conventional cylindrical reactor, it is indispensable to increase the internal volume of the reactor.

The second merit, which can be brought about by passing a gas successively through reaction chambers in accordance with this invention, is concerned with the above-described second structural merit of this invention. Namely, it is relatively easy to draw a high-temperature gas, which has once passed through a bed of a catalyst, out of the pressure-resistant outer shell by means of a tube and then to introduce the gas again into the pressure-resistant outer shell from another location on the surface of the pressure-resistant outer shell. This merit is particularly of value where the feed gas contains a trace amount of a substance which is poisonous to the catalyst. In this case, it is possible to provide in the spherical reactor a reaction chamber which is completely isolated from other reaction chambers. This additional reaction chamber is used as the most upstream-side reaction chamber. Thus, the catalyst packed in this reaction chamber absorbs the catalyst poison and its activity is deteriorated. However, all the catalysts in the downstream-side reaction chambers are protected from the poisonous substance. Thus, the additional reaction chamber can be utilized to prolong the average service life of the entire catalysts without need for replacing the catalysts packed in the downstream-side reaction chambers provided that the catalyst in the most upstream-side reaction chamber is replaced by a fresh one. Furthermore, the second merit permits to successively carry out two or more chemical reactions, which use different catalysts, in a single reactor. A specific example pertaining

to this feature will be described later in this specification.

In the spherical reactor according to this invention, a gas may be passed in either axial direction or radial direction through its reaction chambers. When passing the gas in the axial direction, the gas may be passed from the top to the bottom and from the right to the left or in the opposite directions. On the other hand, when the gas is passed in the radial direction, it is passed from the inside to the outside or *vice versa*. In addition, it is still possible to use the axial flow and radial flow in combination, respectively, for different reaction chambers as desired, for example, by passing the gas in the axial direction through at least one of two or more reaction chambers and in the radial direction through the remaining reaction chambers.

As described above, the internal space of the spherical reactor according to this invention is utilized as reaction chambers, gas flow passages, space for mixing a high temperature gas with a low temperature feed gas, installation space for a heat transfer surface to effect indirect heat exchange, and the like. Designing requirements for these built-in equipments may vary in accordance with a variety of parameters including the type of a reaction which the feed gas undergoes, reaction conditions such as temperature and pressure, type of catalyst, supply rate of the feed gas and the way of removing the reaction heat. Thus, countless embodiments may be contemplated by combining these requirements and parameters in various ways. Several examples of these embodiments will hereinafter be described specifically with reference to the accompanying drawings. It should be borne in mind however that the scope of the present invention is not limited by the following embodiments which are given by way of example.

In Fig. 2, three reaction chambers 31, 32, 33, which are of either intercylinder or cylindrical shape and packed with a catalyst, are arranged coaxially and vertically. A cylindrical protrusion is provided in an upper part of the reactor, which protrusion encloses a shell-and-tube heat exchanger for preheating the feed gas. The reactor is adapted to carry out an exothermic reaction. The feed gas, which has not been preheated to any sufficient extent, is introduced through a pipe 1 and from the lowermost part of a pressure-resistant outer shell 3 into the reactor. It then travels through a space 11 which is in contact with the inner surface of the spherical wall of the pressure-resistant outer shell 3. Thereafter, the feed gas passes *via* a space, which is in contact with the inner surface of the cylindrical protrustion of the pressure-resistant outer shell 3, and reaches an upper space 12 of a heat exchanger 4. Owing to the flow path of the feed gas, the pressure-resistant outer shell is maintained at low temperatures. The feed gas then passes through a number of tubes of the heat exchanger 4 and is subjected to indirect heat exchange with a reaction gas mixture of a high temperature, which has left the reaction chamber 33. After preheating to a desired temperature, the feed gas then reaches a space 13. The feed gas further travels through the internal space of a tube 14 and enters the reaction chamber 31, where a part of the feed gas undergoes a chemical reaction and the temperature of the resultant gas mixture is raised due to reaction heat. This reaction chamber 31 is of an intercylinder shape with its axis passing vertically through the centre of the spherical outer shell. The high temperature gas mixture, which has been subjected partly to the reaction in the reaction chamber 31, enters a space 15 and then passes through an intercylinder space formed subsequent to the space 15 and defined by the reaction chambers 31 and 32. While passing through both of the space 15 and intercylinder space, the high temperature gas mixture is brought into contact with cooling tubes 63, through which a cooling medium is flowing, and cooled to a predetermined temperature. The thus-cooled gas mixture thereafter reaches a space 17. Then, the gas mixture enters the reaction chamber 32 and the reaction of the feed gas further proceeds, thereby raising the temperature of the resulting gas mixture. The gas mixture then reaches a space 19. The reaction chamber 32 is an intercylinder reaction chamber having a vertical axis which passes through the centre of the spherical outer shell. The reaction chamber 32 is coaxial with the reaction chamber 31. Its inner diameter is smaller than that of the reaction chamber 31, but its horizontal cross-sectional area and height may be formed greater than those of the reaction chamber 31. While passing through a space 19 and an intercylinder space 20 provided between the reaction chambers 32 and 33, the gas mixture is brought into contact with cooling tubes 53 through which a cooling medium passes and is cooled to a predetermined temperature. The thus-cooled gas mixture passes through a space 21 and then enters the reaction chamber 33. This reaction chamber 33 has a vertical axis passing through the centre of the spherical outer shell and is a cylindrical reaction chamber provided with a tubular space 23 along its vertical axis. The tubular space 23 is used as a gas flow passage. The chemical reaction of the feed gas is completed in the reaction chamber 33 and the resulting gas mixture, the temperature of which mixture has been raised again, travels through spaces 22, 23 and enters the shell-side of the heat exchanger 4 through an opening 24. As described above, the reaction gas mixture serves to preheat the feed gas in the heat exchanger 4. As a result, it is cooled and flows out of the

reactor through an opening 25 and a gas outlet pipe 2.

On the other hand, the pressure of the cooling medium is adjusted so as to obtain a desired boiling point. It is supplied, as a liquid of approximately its boiling temperature, to distributing headers 51, 61 and introduced into the reactor through tubes 52, 62. It then enters the cooling tubes 53, 63 and subjected to heat exchange with the gas mixtures discharged from the reaction chambers 32, 31 while flowing through the tubes 53, 63, thereby cooling the gas mixtures and being partly evaporated into a vapour phase or a gas-liquid mixed phase. Thereafter, the cooling medium flows through tubes 54, 64 and enters a gas-liquid separator 70, where it is separated into vapour and liquid. The vapour is utilized for given purposes while the liquid is recycled to the distributing headers 51, 61 by gravity or by means of a pump (not illustrated in the drawing).

In the above embodiment, the cooling tubes 53, 63 are both shown as coil-like heat transfer surfaces. They may individually be a coil formed of either a single tube or two or more tubes. These cooling tubes 53 or 63 may be arranged in either a single circle or two or more circles. Namely, the most important thing is to provide a heat transfer area required in accordance with each heat quantity which is subjected to heat exchange. Besides forming the cooling tubes into a coil, they may be formed by arranging a number of substantially vertical tubes in concentric circles as will be described later in this specification.

Fig. 3 illustrates a spherical reactor provided with a reaction chamber resembling a frustum of a cone and another reaction chamber resembling a hollow frustum of a cone (hereinafter called "a truncated intercone shape") and adapted to carry out an exothermic reaction. The reaction gas mixtures are cooled by adding a low temperature feed gas into both reaction chambers. It should be noted that the drawing of Fig. 3 is simplified in order to show the arrangement of the reaction chambers and their configurations. In Fig. 3, numeral 31 is a truncated intercone reaction chamber having a vertical central axis which passes through the centre of the spherical and pressure-resistant outer shell 3. The truncated intercone reaction chamber 31 is packed with a bed of a catalyst. On the other hand, designated at numeral 32 is a reaction chamber resembling a frustum of a cone and having a vertical axis which passes through the centre of the spherical and pressure-resistant outer shell 3. In this embodiment, the feed gas, which has been preheated to the working temperature of the catalyst, enters the reactor through a feed gas inlet 1 formed in an upper part of the reactor. It then passes through two or more gas flow passages 11 provided radially and reaches a toroidal space 12. It then enters the reaction chamber 31. Owing to the contact of the feed gas with the catalyst in the reaction chamber, the reaction is partly proceeded and the gas temperature increases. Within the reaction chamber 31, there is provided a gas cooling system which is constructed of a gas spreader 80 communicating through a tube 71 with the outside of the reactor and adapted to spread and supply the low temperature feed gas evenly in a desired horizontal plane within the reaction chamber. Owing to the supply of a desired amount of the low temperature feed gas by means of the gas cooling system, the gas and catalyst temperatures are prevented from getting excessively high and the reaction is allowed to proceed further. Thus, the reaction gas mixture is again heated and is discharged from the reaction chamber 31 into a space 13, where the reaction gas mixture is supplied with a fresh supply of the low temperature feed gas through another gas cooling system which is formed of a tube 72 extending to the outside of the reactor and a gas spreader 80 communicated with the tube 72. After being mixed with the fresh supply of the feed gas, the resultant gas mixture has been cooled to a given temperature. Thereafter, the thus-cooled gas mixture travels through a space 14 formed between the reaction chambers 31 and 32 and enters *via* a space 15, the reaction chamber 32 where the reaction further proceeds. As a result of a progress of the reaction in the reaction chamber 32, the reaction gas mixture has been heated. Similar to the reaction chamber 31, the thus-heated reaction gas mixture is mixed twice with fresh supplies of the low temperature feed gas which are respectively supplied from two gas spreaders 80 communicated respectively with tubes 73, 74. The reaction gas mixture has thus been cooled and the reaction has thus been completed. The resulting reaction gas mixture travels through a space 16 and reaction gas outlet 2 to the outside of the reactor. It is possible to recover heat from the reaction gas mixture flown out of the reactor, for example, by a waste-heat boiler. This part is however unillustrated in the drawing. Since the additional supply of the low temperature feed gas is effected for cooling the reaction gas mixture in each of the reaction chambers and in a space between both of the reaction chambers in this embodiment, more gas tends to pass as reaction chambers become closer to the downstream end. However, by using reaction chambers resembling a frustum of a cone or a truncated intercone shape and causing a gas to flow in a direction from the top of each chamber toward its bottom, the cross-sectional area of the gas flow passage increases as the gas flows downwardly. This prevents the flow velocity of the gas from getting faster in the reaction chamber and makes the pressure loss, which occurs due to the passage of the

gas, relatively small, thereby avoiding the conversion rate from lowering due to an increase in space velocity.

Similarly, the cross-sectional area of the gas flow passage may be increased as reaction chambers become closer to the downstream end. This is effective to lower the flow velocity of the gas and thus to obtain further effects owing to a reduced pressure loss. In addition, the space velocity may be maintained at a suitable level by increasing the packed amount of the catalyst as reaction chambers become closer to the downstream end.

In Fig. 4, there are provided a cylindrical reaction chamber 31 and another reaction chamber 32 for carrying out an exothermic reaction. The reaction chamber 32 is formed using a toroidal space defined by the circumferential wall of the former reaction chamber 31 and the spherical wall of the pressure-resistant outer shell 3. The reaction chambers 31, 32 share the same central axis. The gas is cooled, similar to the reactor shown in Fig. 3, by adding a low-temperature feed gas. However, the preheating of the feed gas and cooling of a reaction gas mixture, which has not completely undergone the reaction, are effected in a shell-and-tube heat exchanger 4 provided in an upper part of the spherical outer shell 3 by subjecting both gases to indirect heat exchange. In the drawing, the illustration is simplified in order to show the arrangement of the reaction chambers and their configurations. In Fig. 4, numeral 31 indicates the cylindrical reaction chamber which has a vertical central axis passing through the centre of the spherical and pressure-resistant outer shell 3. On the other hand, numeral 32 is the toroidal reaction chamber which has a vertical central axis passing through the centre of the pressure-resistant outer shell 3 and is formed by the spherical wall of the outer shell 3 and a part of the outer side wall of the reaction chamber 31. In the present embodiment, the feed gas, which has not been preheated to a sufficient level, enters through a tube 1, travels *via* a space 11 formed in an upper part of a heat exchanger 4, and passes through a number of tubes of the heat exchanger 4. Here, the feed gas is subjected to indirect heat exchange with a reaction gas mixture of a high temperature, which reaction gas mixture has been discharged from the reaction chamber 31, and is preheated to a desired temperature. Then, the thus-preheated feed gas enters a space 12 and then the reaction chamber 31, where it is brought into contact with a catalyst and its reaction proceeds to a certain extent. Thus, the temperature of the resulting gas mixture has been raised. In the reaction chamber 31, there is provided a gas cooling system formed of a gas spreader 80 which is communicated through a tube 71 with the outside of the reactor and is adapted to spread a fresh supply of the low temperature feed gas evenly in a given horizontal plane within the reaction chamber. Owing to the provision of the gas cooling system, the temperatures of the reaction gas mixture and catalyst are prevented from getting excessively high by the addition of a desired amount of the low temperature feed gas. Thus, the reaction is allowed to proceed further and the resulting reaction gas mixture is again heated. The thus-heated reaction gas mixture is mixed twice with fresh supplies of the low temperature feed gas charged through two spreaders 80 which are respectively communicated with tubes 72, 73 and is thus cooled. The thus-cooled reaction gas mixture thereafter leaves the reaction chamber 31, passes through spaces 13, 14, and enters the shell side of the heat exchanger 4 *via* an opening 15. Here, the reaction gas mixture serves to preheat the feed gas and its temperature is correspondingly lowered to a predetermined level. The thus-cooled reaction gas mixture further travels through spaces 16, 17, 18, and enters the reaction chamber 32. Its reaction further proceeds in the reaction chamber 32 and the resulting reaction gas mixture flows out of the reactor through a space 19 and outlet 2. In Fig. 4, the cross-sectional area of the gas flow passage in the reaction chamber 31 is identical to that in the reaction chamber 32. However, in the structure shown in Fig. 4, it is possible to form each of the reaction chambers into a frustum of a cone as shown in Fig. 3 or a multi-staged cylindrical shape so as to change the cross-sectional area of its gas flow passage, thereby reducing the pressure loss and suitably controlling the reaction conditions.

Fig. 5 illustrates a still further embodiment of this invention, in which the gas is caused to flow in the radial direction through an intercylinder reaction chamber 31 and another intercylinder reaction chamber 32 provided inside the reaction chamber 31 so as to undergo an exothermic reaction. Within the intercylinder reaction chamber 32, a plurality of cooling tubes 53 are vertically arranged in one or more concentric circles for allowing a cooling medium to pass therethrough. The feed gas, which has not been preheated, enters the reactor through a gas inlet 1 provided in a lower part of the reactor. It then passes through a space 11 formed along the inner surface of the spherical and pressure-resistant outer shell 3. It thereafter flows into an upper compartment 12 of a shell-and-tube heat exchanger 4 which is provided in the central space of the reaction chamber 32. The feed gas passes through a plurality of tubes of the heat exchanger 4. Here, it is subjected to heat exchange with a reaction gas mixture of a high temperature which has completed the reaction, thereby preheated to a predetermined temperature. Thereafter, the thus-pre-

heated feed gas enters a lower compartment 13 of the heat exchanger 4. The thus-pre-heated feed gas then enters a peripheral equalizer space 15 through the space 14 within radially-arranged connector tubes, from which it flows in substantially horizontal and radial direction through the interior of the reaction chamber 31, in other words, from the outer circumferential part of the intercylinder reaction chamber 31 toward its central axis which extends through the centre of the spherical and pressure-resistant outer shell. Since the feed gas is brought into contact with the catalyst in the reaction chamber 31, an exothermic reaction is induced and the resulting gas mixture is heated. Similar to the embodiment shown in Fig. 3, a fresh supply of the low temperature feed gas is introduced through a tube 71 from the outside of the reactor and dispersed by a gas spreader 80 evenly in a cylindrical plane coaxial with the reaction chamber 31, thereby preventing the reacting gas mixture from getting too hot. The reaction gas mixture then flows out of the reaction chamber 31 and flows into an inter-cylinder equalizer space 16 provided between the reaction chamber 31 and the intercylinder reaction chamber 32 disposed inside the reaction chamber 31. After tentatively held in the equalizer space 16, the reaction gas mixture flows into the intercylinder reaction chamber 32 in substantially horizontal and radial direction, namely, toward the central axis of the reactor. The reaction chamber 32 is longer in the vertical direction, compared with the reaction chamber 31. The reaction is again allowed to proceed. As mentioned above, the reaction chamber 32 is provided with the plurality of cooling tubes 53 arranged in concentric circles. Owing to a cooling medium passing through the cooling tubes 53, the reaction heat developed in the reaction chamber 32 is suitably removed, thereby successfully preventing the internal temperature of the reaction chamber 32 from becoming too high. In this embodiment, the reaction chamber 32 is equipped with the above-described cooling system. This is because such a cooling system is capable of exhibiting a very big cooling power when a cooling medium, which for example has a desired boiling point at a suitable pressure, is caused to pass through the cooling tubes 53 while boiling. Furthermore, by controlling the density of the arrangement of these cooling tubes in the reaction chamber 32, it is also possible to bring the temperature distributions in the catalyst bed and gas flow along the flowing direction of the gas through the reaction chamber 32 (namely, in the radial direction) into conformity with desired temperature distributions which have been preset. It is important to suitably preset the gas temperature distribution along the flowing direction of a gas in a catalyst bed in order to prevent the concentration and yield of the reaction product from lowering due to a lowered reachable upper limit of the reaction, which is caused by chemical equilibratory phenomena frequently observed in exothermic reactions when gas temperatures are raised, or to obtain a given quantity of a reaction product while minimizing the amount of the catalyst required therefor. The reaction gas mixture, which has radially passed through the reaction chamber 32 after completing its reaction and is still hot, flows out into an intercylinder space 17 between the heat exchanger 4 and reaction chamber 32. Afer that, it flows into the shell side of the heat exchanger 4 from a lower part thereof and serves, as described above, to preheat the feed gas. Then, it flows out of the reactor through the gas outlet 2.

On the other hand, the cooling medium is introduced at a desired pressure as a liquid of its boiling point into the reactor from the outside of the reactor by means of a tube 51. Thereafter, it is distributed into a plurality of cooling tubes 53 by one or more toroidal distributing headers 52 which are in communication with the tube 51. It then passes as ascending flows through the cooling tubes 53 while boiling and gathers into one or more toroidal collecting tubes 54. It then flows *via* a tube 55 into a gas-liquid separator 70 provided outside the reactor. In the gas-liquid separator 70, the cooling medium is separated into liquid and vapour. The vapour is guided through a tube 56 for a given purpose while the liquid is recycled, without being cooled, to the tube 51 by its gravity or by means of a pump. This part is not shown in the drawing. In the illustrated reactor, the weights of the reaction chambers are suspended by means of turnbuckles 91 provided at upper parts of the reactor. This structure is more advantageous from the viewpoints of thermal stress and strength, compared with supporting these reaction chambers at their bottoms.

Fig. 6 illustrates a still further embodiment of this invention, which makes use of heat exchangers installed outside the reactor. In the illustrated embodiment, the reactor is equipped with two intercylinder reaction chambers 31, 32 and one cylindrical reaction chamber 33, all of which chambers have a common axis extending along a line that passes through the centre of the reactor. The reaction chambers 31, 32, 33 are respectively divided into two equal halves which are respectively isolated in two hemispheres divided by a plane extending through the centre of the spherical reactor and perpendicular to the above-described common axis. The thus-divided independent reaction chambers are respectively indicated by numerals 31*a* and 31*b*; 32*a* and 32*b*; and 33*a* and 33*b*. Toroidal spaces 13 and 17 are respectively formed as gas flow passages between the reaction

chambers 31*a* and 31*b* and between the reaction chambers 32*a* and 32*b*. On the other hand, a disc-shaped space 20 is interposed between the reaction chambers 33*a* and 33*b*. A preheated feed gas is introduced into the reactor through the inlet 1 of a double-walled tube and, first of all, enters a space 11. From this space 11, the flow of the feed gas is divided approximately into two equal portions. One of the flows, namely, the flow *a* passes through a clearance formed between the reaction chamber 31*a* and the inner surface of the spherical and pressure-resistant outer shell as indicated by the arrow and flows, through a toroidal space 12*a*, into the reaction chamber 31*a*. The other flow *b* passes through a clearance formed between the reaction chamber 31*b* and the inner surface of the spherical and pressure-resistant outer shell as indicated by the arrow and flows, *via* a toroidal space 12*b*, into the reaction chamber 31*b*. The feed gas, which has entered the reaction chambers 31*a* and 31*b*, is caused to undergo a chemical reaction in both of the reaction chambers. The resulting reaction gas mixtures, which have been heated where the reaction is an exothermic reaction or have been cooled where the reaction is an endothermic reaction, are then merged and caused to flow into the space 13. The thus-merged reaction gas mixture passes through the space 14 and flows into an indirect heat exchanger 8 provided outside the reactor. Its temperature is then adjusted owing to a heat exchange which occurs between the reaction gas mixture and a fluid of a desired temperature, which fluid flows in through a tube 81*a* and flows out through a tube 81*b*. Namely, the reaction gas mixture is cooled where its temperature, after being merged in the space 13, is excessively higher than an appropriate temperature for a catalyst bed disposed right after the heat exchanger 8. On the contrary, where its temperature is excessively low, it is heated to a desired temperature. The resulting stream of the reaction gas mixture, the temperature of which mixture has been adjusted, is again divided into approximately two equal halves. One of the flows, namely, the flow *a* passes through a tube 15*a* and flows, *via* a toroidal space 16*a*, into the reaction chamber 32*a*, while the other flow *b* passes through a tube 15*b* and flows, *via* a toroidal space 16*b*, into the reaction chamber 32*b*. The chemical reaction is again induced in each of the reaction chambers 32*a* and 32*b*. The temperature of the reaction gas mixture increases when the reaction is an exothermic reaction but drops when the reaction is an endothermic reaction. After passing through the reaction chambers 32*a* and 32*b*, the resultant reaction gas mixtures flow into the toroidal space 17, where they are combined together. The thus-merged reaction gas mixture flows into another heat exchanger 9 through a space 18. In the heat exchanger 9, the temperature of the reaction gas mixture is adjusted in the same manner as described above and then divided into approximately two equal halves, which flow, respectively, *via* tubes 19*a* and 19*b* into the reaction chambers 33*a* and 33*b*. The reaction gas mixtures, which have respectively flown into the reaction chambers 33*a* and 33*b*, undergo a chemical reaction again, thereby changing their temperatures. After completion of the chemical reaction, they are again merged in the space 20 and then discharged from the reactor through an outlet 2 for the reaction gas mixture. This outlet 2 is provided through the pressure-resistant outer shell 3 in a direction perpendicular to the drawing sheet. The gas, which has flown out of the reactor through the outlet 2, is still hot and heat may be recovered from the thus-discharged gas. However, this part is not shown in the drawing. In the illustrated embodiment, the pressure loss is reduced by dividing each of the intercylinder and cylindrical reaction chambers into two halves and using them in parallel. Owing to the above constitution, the pressure-resistant outer shell can be maintained at relatively lower temperatures even if it is used for an exothermic reaction.

In the above embodiment, the central axis common to each of the reaction chambers extends vertically. The reactor may however be used without encountering any problems or inconveniences even it this common axis extends horizontally or aslant, provided that the position of each gas-permeable catalyst support is suitably adjusted. The chemical reaction may be either exothermic or endothermic reaction as described above. In addition, the same catalyst may be used as catalysts to be packed in the reaction chambers 31, 32 and 33 in order to carry out the same chemical reaction in each of the reaction chambers. It is also possible, also for carrying out the same chemical reaction in each of the reaction chambers, to use different catalysts which are adapted to induce the same chemical reaction but are different in temperature characteristics, optimum space velocity and the like. It is also possible to carry out three different chemical reactions, which are required to effect as two-stage or three-stage chemical reactions using two or three reactors when conventional reactors are relied upon, substantially as a single stage reaction, as described above, by packing different catalysts in the reaction chambers 31, 32, 33 to induce the different chemical reactions therein and causing the feed gas and reaction gas mixtures, the temperatures of which gas and gas mixtures have been adjusted to temperatures suitable for the different catalysts respectively, to pass successively through the beds of the different catalysts. The different chemical reactions may include one or more exothermic reactions and endothermic reactions in

combination. Upon carrying out such different chemical reactions, it is preferred to use a heat-insulative material for each partition wall interposed between each two adjacent reaction chambers where the temperature of one reaction chamber is considerably different from that of another reaction chamber. In addition, it is also possible to carry out in the reaction chamber 33 a chemical reaction, which is different from that carried out in the other two reaction chambers if desired, by additionally supplying through an additional gas feeding port 10 another feed gas which is different from the feed gas supplied through the gas inlet 1. On the other hand, it may be possible to interpose, between the space 18 and heat exchanger 9, means for separating and removing a certain reaction product from those occurred in the reaction chambers 31, 32.

As an example of carrying out such different chemical reactions, may be mentioned to synthesize methanol from hydrogen gas and carbon oxide and subsequently to obtain synthetically hydrocarbons from the thus-synthesized methanol. Namely, a high pressure feed gas rich in hydrogen gas and the carbon oxide is supplied through the feed gas inlet 1. Using a known methanol synthesis catalyst (for example, that containing chromium, copper, zinc and/or the like) in both of the reaction chambers 31 and 32, the feed gas is converted at the pressure of 100 kg/cm^2 and temperature of about 300°C into a gas mixture consisting of methanol, hydrogen gas and the carbon oxide. By causing the thus-obtained gas mixture without separating methanol from the unreacted raw materials but after adjusting its temperature to 350°C or so by the heat exchanger 9 into the reaction chamber 33 which contains a zeolite catalyst, methanol can be converted into hydrocarbons. Unreacted hydrogen gas and carbon oxide are, after separating methanol and hydrocarbons out from the reaction gas mixture, are recirculated to the feed gas inlet 1, while methanol, which has not been converted into hydrocarbons, is returned to the additional raw material feeding port 10 and then introduced back into the reaction chamber 33 as vapour or liquid after separating it from hydrocarbons.

A number of merits can be brought about, as mentioned above, by the reactor according to this invention and the method of its use. In summary, the first merit relates to the reactor. Namely, the present invention enables to make the wall thickness of the pressure-resistant outer shell of the reactor and correspondingly to reduce the weight of the reactor while still allowing to pack the same amount of a catalyst. Owing to this merit, it is possible to save the steel material for the fabrication of a reactor and to fabricate the reactor *per se* with a lower cost. In addition, it is also feasible to make the foundation, support, pipings and the like, which are required for the installation of the reactor according to this invention, smaller or shorter. Accordingly, the overall construction cost of the reactor can be cut down. This merit becomes remarkable, particularly, the reactor is a high pressure reactor of a large capacity. The second merit of this invention pertains to the method of carrying out a reaction. Since the reactor according to this invention is equipped with two or more of the above-described reaction chambers, which are of a cylindrical, intercylinder, truncated conical or truncated intercone shape and are different in the cross sectional area perpendicular to the passing direction of a gas therethrough and the amount of a catalyst packed therein, these reaction chambers having different configurations may be suitably designed at will in accordance with the nature of chemical reactions carried out therein and the characteristics of catalyst packed therein. Namely, when carrying out a single chemical reaction at the same space velocity (particularly in a large reactor), a cylindrical reactor is required to be very thin but very long. If a gas is passed in the axial direction of the cylinder, the channelling of the gas in the catalyst bed can be minimized. However, this leads to a very fast flow velocity, resulting in an increased pressure loss. On the contrary, when the gas is passed in the radial direction with a view toward making the pressure loss smaller, the flow velocity becomes uneven along the long axis of the cylindrical reactor. However, when a spherical reactor is employed as in the present invention, each of its internal reaction chambers is formed into a shape having a small diameter-to-height ratio, in other words, is thick and short. Accordingly, the reactor according to this invention does not develop the channelling problem and can be used as a reactor featuring a small pressure loss.

The above merit can also bring about a favourable effect for prolonging the service life of a catalyst. The prolonged service life of a catalyst has been derived from two reasons which are different in principle. The first reason may be explained, for example, with reference to a reaction accompanied by a generation of large heat such as the synthesis of methanol from hydrogen gas and carbon oxide. In reactions of the above sort, when a gas is caused to pass through each of catalyst beds at a uniform flow velocity which is the average flow velocity of the flow velocities suitable respectively to all the catalysts, a violent exothermic reaction generally takes place at the catalyst to which the feed gas is first brought into contact since the concentration of a reaction product is still low there. Such a violent exothermic reaction makes the catalyst excessively hot, thereby developing the thermal deterioration phenomenon as to

the activity of the catalyst. This phenomenon can be avoided by passing the gas at a high velocity through the catalyst bed. If one wants to use a cylindrical reactor for carrying out a reaction of the above-described type without developing the phenomenon, it becomes necessary to increase the length of the reactor when the gas is fed as an axial flow or to leave one or more useless spaces in the reactor when the gas is passed as a radial flow. If the length of the reactor is increased as mentioned above, the pressure loss will become greater. Accordingly, neither the former nor the latter measure is practical. Contrary to this, it is easy to form small short and thick reaction chambers in a reactor without increasing the useless space therein, when the present invention is incorporated. Therefore, the above-described phenomenon can be successfully avoided. The second reason for the prolonged service life of the catalyst is found under the following situation. Where a feed gas contains a trace amount of a substance poisonous to the catalyst as described above, a small space enclosing a bed of the catalyst (i.e., a reaction chamber), to which the feed gas is brought into contact at first, may be formed in such a structure that it is sufficiently isolated from other reaction chambers and spaces for a heat exchanger and cooling system in the reactor. If these reaction chambers are connected by means of pipings which are provided outside the spherical reactor and equipped with valves, it becomes possible, when the catalyst contacted first with the feed gas has been poisoned, to isolate the reaction chamber containing the thus-poisoned catalyst from the other reaction chambers by suitably operating the valves and to replace the thus-poisoned catalyst with a fresh supply of the catalyst, thereby prolonging the service life of the remaining catalyst, i.e., the majority of the catalyst.

The third merit of this invention relates to the utilization method of the reactor according to this invention. As has already been mentioned in the description of the embodiment shown in Fig. 6, the reactor of this invention can be used to carry out two or more different chemical reactions which take place at the same pressure and the same or different temperatures. Although the reactor according to this invention may be used as a reactor for parallelly carrying out two or more chemical reactions which are not correlated, the use of the reactor according to this invention generally makes it possible to carry out two or more chemical reactions substantially as a single step reaction so as to obtain a final reaction product from raw materials by effecting the two or more chemical reactions successively one after another. Thus, the method of this invention is suitable for decreasing the number of reactors.

The reactor according to this invention is suitable as a reactor for obtaining at pressures above 5 kg/cm^2 a gaseous reaction product from gaseous raw materials and the above-described merits become greater as the reaction pressure increases and the internal volume of each reactor becomes larger. As chemical reactions preferably carried out using the reactor according to this invention, the following reactions may be mentioned: synthesis reaction of ammonia from a gas containing hydrogen gas and nitrogen gas; synthesis reactions of aliphatic monohydric alcohols such as methanol, ethanol, propanols and butanols from a gas containing hydrogen gas and a carbon oxide; direct synthesis reaction of hydrocarbons from a gas containing hydrogen gas and a carbon oxide (i.e., the so-called Fischer-Tropsch synthesis); synthesis processes including synthesizing aliphatic monohydric alcohols from a gas containing hydrogen gas and carbon oxide and then converting the thus-obtained aliphatic monohydric alcohols into hydrocarbons; reaction for converting a gas containing steam and carbon monoxide into another gas containing hydrogen gas and carbon dioxide; reaction for converting a gas containing one or more hydrocarbons such as methane and/or naphtha and steam into another gas containing hydrogen gas and a carbon oxide; and reaction for synthesizing saturated hydrocarbons from a gas containing one or more unsaturated hydrogen gas and hydrogen gas. The above reactions and processes are principal examples.

When carrying out these chemical reactions, reaction conditions such as temperature, pressure and space velocity vary from one reaction to another in accordance with the kinds of reactions, the characteristics of catalysts to be used, etc. However, it is generally feasible to carry out such reactions under substantially the same conditions as those well-known for conventional cylindrical reactors.

Furthermore, known gas spreaders, heat exchangers and gas-permeable catalyst supports may be used as the spreaders for supplying a low temperature feed gas upon cooling the catalysts and gases in the reactor according to this invention, the various heat exchangers and gas-permeable catalyst supports. As cooling media useful for cooling catalysts and reaction gas mixtures, which have not gotten through their reactions, in accordance with the indirect heat exchanging method, may be mentioned as preferred examples water, the pressure of which has been adjusted so that it boils at a desired temperature; one of hydrocarbons which are liquid at room temperature, or a mixture of at least two of such hydrocarbons; a heat transfer medium such as "DOWNTHERM" (trade mark; product of the Dow Chemical Company). However, gaseous cooling media or other cooling

liquid media may be sufficiently employed in some instances.

Having now fully described the invention, it will be apparent to one of ordinary skill in the art that many changes and modifications can be made thereto without departing from the spirit or scope of the invention as set forth herein.

CLAIMS

1. A reactor adapted to bring a feed gas into contact with a fixed bed of a granular catalyst under elevated pressures so that the feed gas is caused to undergo a chemical reaction and a gaseous reaction product is thus obtained, said reactor comprising a pressure-resistant outer shell and at least two reaction chambers enclosed within the outer shell, the outer shell being either (a) a sphere or a spheroid formed of a cylinder having a length equal to or shorter than the three quarters of its diameter and hemispherical end plates having the same diameter as the cylinder and provided at both ends of the cylinder or (b) another sphere or spheroid having the same configurations as the former sphere or spheroid and provided with at least one cylindrical protrusion which covers a quarter of the overall surface area of the latter sphere or spheroid or less; and the reaction chambers being of cylindrical, intercylinder, truncated conical and/or truncated intercone shapes and/or shapes defined respectively between parts or the entire parts of the outer surfaces of the cylindrical, intercylinder, truncated conical or truncated intercone shapes and the inner surface of the outer shell and having annular cross-sections when taken along planes perpendicular to the axes thereof and being arranged in such relationship that the reaction chamber having a smaller outer diameter is present inside the reaction chamber having a greater inner diameter.

2. A reactor according to claim 1, wherein said reactor further comprises at least one bent tube extending through the spherical wall or spheroidal wall of the pressure resistant outer shell and a division wall dividing the interior space of the reactor so as to communicate the thus-divided interior space of the reactor with the exterior of the reactor; and the bent tube is positioned, between the outer shell and division wall, within a space in the proximity of the inner surface of the spherical or spheroidal wall of the outer shell.

3. A reactor according to claim 1 or 2, wherein (a) the reaction chambers are individually of an intercylinder shape and disposed upright, (b) said reactor further comprises an equalizer space provided between each two adjacent reaction chambers and (c) a multiplicity of cooling tubes are arranged within at least one of the reaction chambers, in at least two concentric circles and parallel to the axis of the reaction chamber; whereby causing the feed gas to flow radially and successively through each of the reaction chambers and allowing a cooling medium to flow upwardly at a desired pressure as a boiling gas-liquid mixed phase through the cooling tubes.

4. A reactor according to any one of claims 1 to 3, wherein the outer shell is a sphere or spheroid provided with at least one cylindrical protrusion; and within at least one of said cylindrical protrusions an indirect heat exchanger is provided so as to preheat the feed gas with a high-temperature gas occurred in the course of the reaction or after completion of the reaction.

5. A method of bringing a feed gas into contact with a fixed bed of a granular catalyst under elevated pressures so that the feed gas is caused to undergo a chemical reaction and a gaseous reaction product is thus obtained, characterised in that the reaction is proceeded by using the reactor according to claim 1 and causing the feed gas to pass successively through at least two reaction chambers in the reactor.

6. A reactor adapted to bring a feed gas into contact with a fixed bed of a granular catalyst under elevated pressures so that the feed gas is caused to undergo a chemical reaction and a gaseous reaction product is thus obtained, substantially as hereinbefore described with reference to, and as shown in, the accompanying drawings.

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